

Unusual criticality of $\text{Cu}_2\text{Te}_2\text{O}_5\text{Br}_2$ under pressure

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We present measurements of the magnetic susceptibility $\chi(T)$ on $\text{Cu}_2\text{Te}_2\text{O}_5\text{Br}_2$ under externally applied pressure. From our data we extract the pressure response of the antiferromagnetic phase transition at $T_0 = 11.6$ K and of the overall magnetic coupling strength. Our experiments indicate that with pressure the overall magnetic coupling strength increases by about 25% with applied pressure of only ~ 8 kbar. In contrast, the phase transition temperature T_0 is significantly suppressed and not observable anymore at a pressure of already 8.2 kbar.

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The spin-tetrahedra system $\text{Cu}_2\text{Te}_2\text{O}_5\text{Br}_2$ [1] belongs to a class of quantum magnets which has been in the focus of intense research efforts recently [2, 3, 4, 5, 6]. Here, the presence of a spin gap through dimerization for a quantum magnet does not lead to a non-magnetic singlet ground state. Instead, based on thermodynamic and spectroscopic techniques an unusual magnetic ground state has been evidenced. Tetragonal $\text{Cu}_2\text{Te}_2\text{O}_5\text{Br}_2$ contains clusters of Cu^{2+} with $S = 1/2$ in a distorted square planar CuO_3Br coordination (Fig. 1). These tetrahedra form weakly coupled sheets within the crystallographic a - b -plane. Therefore, this system is ideal to study the interplay between the spin frustration of a tetrahedron with localized low energy excitations and the tendency for a more collective magnetism induced by inter-tetrahedra couplings.

The thermodynamic properties of $\text{Cu}_2\text{Te}_2\text{O}_5\text{Br}_2$ are ascribed to two magnetic couplings within the tetrahedra, with the competing exchange constants J_1 and J_2 [2], and an inter-tetrahedra coupling J_c [9]. As result of the coupling, the system undergoes a phase transition at $T_0 = 11.6$ K. Neutron powder diffraction of $\text{Cu}_2\text{Te}_2\text{O}_5\text{Br}_2$ reveals an antiferromagnetically ordered state with a strongly reduced magnetic moment of $0.51(5)\mu_B/\text{Cu}^{2+}$ below T_0 [5]. On a microscopic level the cause for the phase transition has not unambiguously been resolved [7, 8, 9, 10, 11, 12]. In particular, with the existence of low lying excitations a magnetically ordered state close to quantum criticality has been discussed.

Pressure experiments have proven to be a particu-

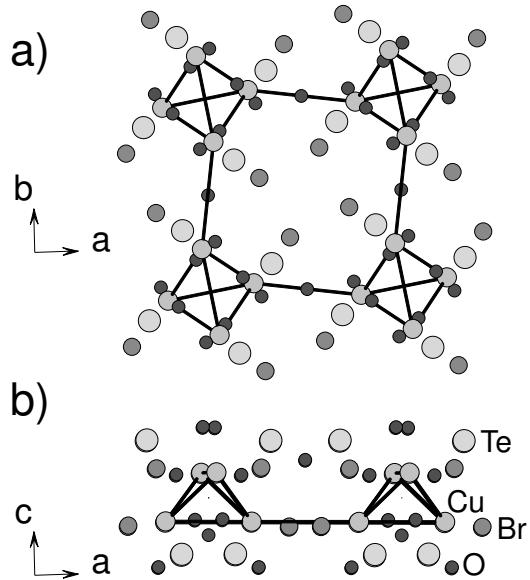


FIG. 1: A view of the crystal structure of $\text{Cu}_2\text{Te}_2\text{O}_5\text{Br}_2$ onto the crystallographic a - b (a) and a - c plane (b) as illustration for the planar arrangement of the Cu^{2+} tetrahedra.

larly useful tool to study quantum critical behavior. Therefore, in this work we present a pressure study on $\text{Cu}_2\text{Te}_2\text{O}_5\text{Br}_2$. For our experiments we used a CuBe pressure cell in a commercial SQUID magnetometer to measure $\chi(T)$ at pressures up to 8.2 kbar and in external fields up to 5 T for temperatures 2 - 40 K. A powder sample, which has been prepared as described in Ref. [1], was pressed together with GE-Varnish into a pellet,

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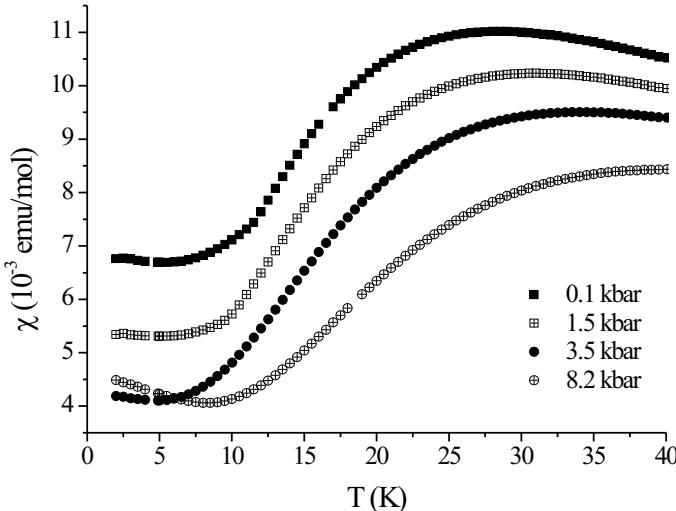


FIG. 2: The magnetic susceptibility $\chi(T)$ of $\text{Cu}_2\text{Te}_2\text{O}_5\text{Br}_2$ as function of pressure, measured in an external field of 5 T.

which was placed in the middle of a teflon tube. The tube was filled with a hydraulic pressure medium (FC-77) and was loaded into the CuBe pressure cell. In Fig. 2 we plot a set of representative measurements on $\text{Cu}_2\text{Te}_2\text{O}_5\text{Br}_2$ with our cell for pressures up to 8.2 kbar.

According to ambient pressure experiments [2], the magnetic susceptibility $\chi(T)$ exhibits a broad maximum at about $T_{Max} = 30$ K. This susceptibility maximum represents a measure for the overall magnetic coupling strength, *viz.*, the size of J_1 and J_2 . Below T_{Max} a strong reduction of χ occurs, as is typical for the onset of antiferromagnetic correlations. The ordering temperature T_0 is identified as a step in the temperature derivative $\partial\chi/\partial T$.

Our data at 0.1 kbar closely resemble the ambient pressure behavior from Ref.[2]. Since a step in $\partial\chi/\partial T$ corresponds to a maximum in $\partial^2\chi/\partial T^2$, we determine T_0 at 0.1 kbar from the latter quantity to 11.6 K, in good agreement with Ref.[2] (Fig.3).

With increasing pressure the phase transition temperature T_0 decreases, and we obtain $T_0 = 9.8$ K at 1.5 kbar (Fig.3). At 3.5 kbar the maximum in $\partial^2\chi/\partial T^2$ has shifted to 5 K. However, since for both the data taken at 1.5 kbar and 8.2 kbar a similar, but much smaller maximum in $\partial^2\chi/\partial T^2$ appears, this feature at 5 K might possibly be induced by a changing cooling mode of the SQUID in this temperature range.

Alternatively, it could be argued that for the 3.5 kbar measurement the broad anomaly underlying the peak at 5 K represents a remnant of the antiferromagnetic ordering. In that case the data would indicate a range of T_0 between 5 and 8 K. Still, the observation of similar broad anomalies for the higher pressure experiments seems to speak against such interpretation.

In Fig.4 we summarize the pressure dependence of T_0 . The error bar at 3.5 kbar reflects the uncertainty about

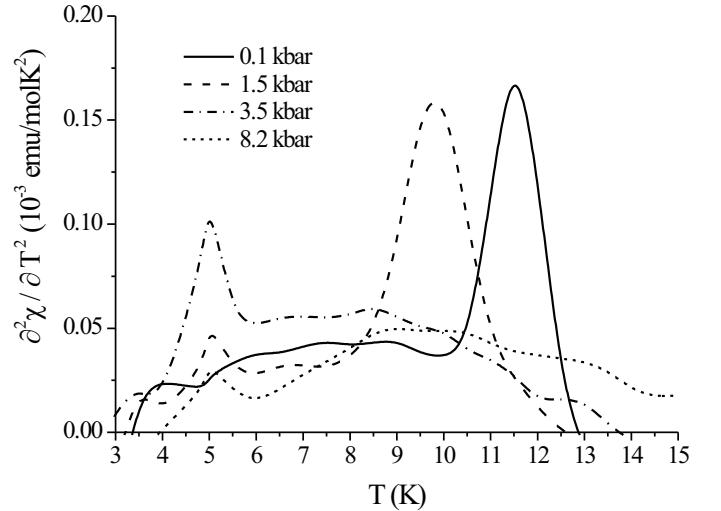


FIG. 3: The second derivative $\partial^2\chi/\partial T^2$ of $\text{Cu}_2\text{Te}_2\text{O}_5\text{Br}_2$ as function of pressure in an applied field of 5 T.

the determination of T_0 at this pressure. Altogether, the data suggest a suppression of T_0 in the range 5-8 kbar. This statement is supported by the absence of any clear signature of magnetic ordering for the measurement at 8.2 kbar. Hence, our experiments indicate that $\text{Cu}_2\text{Te}_2\text{O}_5\text{Br}_2$ is situated in the proximity to a nonmagnetic phase. As the constituting unit is a tetrahedron with antiferromagnetic exchange interaction this phase is suggested to be identical with a short range correlated singlet phase. However also other scenarios have been put forward based on theoretical arguments [11]. Two experimental finding shine further light on the peculiarity of $\text{Cu}_2\text{Te}_2\text{O}_5\text{Br}_2$. First is the observation of a related instability with a much higher $T_0=18.2$ K in $\text{Cu}_2\text{Te}_2\text{O}_5\text{Cl}_2$ that has a 7% smaller unit cell volume [2]. This compound also has a completely different low energy excitation spectrum in light scattering experiments [3]. The second observation is the evidence for an incommensurate ordering vector of $\text{Cu}_2\text{Te}_2\text{O}_5\text{Br}_2$ for $T < T_0$ [5]. Both experimental results imply that the ordering temperature is not only given by a mean field-like inter-tetrahedra coupling and that some additional effect, most probably some antisymmetric Dzyaloshinskii-Moriya (DM) interaction plays some role to establish long range ordering.

Moreover, from $\chi(T)$ we derive the pressure dependence of the susceptibility maximum. It increases from $T_{Max} = 28.5$ K (determined via $\partial\chi/\partial T = 0$) at 0.1 kbar applied pressure to $T_{Max} = 40$ K at 8.2 kbar (Fig.4). This increase indicates a very substantial strengthening of the intra-tetrahedra coupling with applied pressure, yielding an enhancement of 25% at highest applied pressure.

The contrasting pressure response of T_0 and T_{Max} is very unusual and likely reflects competing energy scales. If the ordering temperature T_0 would be only controlled by the overall magnetic coupling strength, we would ex-

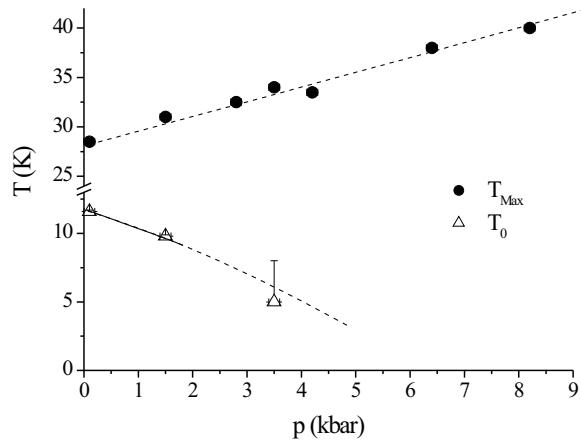


FIG. 4: The pressure dependence of T_{Max} and T_0 in an external field of 5 T. Lines are guides to the eye.

pect an increase of T_0 with T_{Max} . Therefore, the decreasing T_0 possibly is the result of enhanced frustra-

tion J_1/J_2 on the tetrahedra. Another scenario would be a weakened inter-tetrahedra coupling J_c with pressure. This however seems unlikely as under pressure the inter-tetrahedra distance decreases which in the absence of structural symmetry modifications should lead to an increase of J_c .

In summary, we have performed a pressure study on the susceptibility of $\text{Cu}_2\text{Te}_2\text{O}_5\text{Br}_2$. We have determined the pressure response of the antiferromagnetic phase transition temperature T_0 and the overall magnetic coupling strength. While we find a strengthening of the magnetic coupling with pressure, attributed to intra-tetrahedra exchange paths, antiferromagnetic order is rapidly suppressed. Tentatively, we relate this behavior to an enhancement of frustration or a weakening of anti-symmetric interactions in the system. However, to weight such scenarios $\text{Cu}_2\text{Te}_2\text{O}_5\text{Br}_2$ additional thermodynamic and spectroscopic pressure experiments to higher pressure, structural studies under pressure and following theoretical investigations will be necessary. Such work is in preparation.

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